

# Mercury concentrations and pools in four Sierra Nevada forest sites, and relationships to organic carbon and nitrogen

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Received: 17 November 2008 – Published in Biogeosciences Discuss.: 9 February 2009

Revised: 27 April 2009 – Accepted: 27 April 2009 – Published: 6 May 2009

**Abstract.** This study presents data on mercury (Hg) concentrations, stoichiometric relations to carbon (C) and nitrogen (N), and Hg pool sizes in four Sierra Nevada forest sites of similar exposure and precipitation regimes, and hence similar atmospheric deposition, to evaluate how ecosystem parameters control Hg retention in ecosystems. In all four sites, the largest amounts of Hg reside in soils which account for 94–98% of ecosystem pools. Hg concentrations and Hg/C ratios increase in the following order: Green Needles/Leaves < Dry Needles/Leaves < Oi litter < Oe litter < Oa litter. Stoichiometric relations show negative correlations between Hg and C ( $r^2=0.58$ ) and N and C ( $r^2=0.64$ ) in decomposing litter, but a positive correlation between litter Hg and N ( $r^2=0.70$ ). These inverse relations may reflect preferential retention of N and Hg over C during decomposition, or may be due to older age of decomposed litter layers which are exposed to longer-term atmospheric Hg deposition in the field. The results indicate that litter Hg levels depend on decomposition stage and may not follow generally observed positive relationships between Hg and organic C.

Mineral soil layers show strong positive correlations of Hg to C across all sites and soil horizons ( $r^2=0.83$ ), but Hg concentrations are even more closely related to N with a similar slope to that observed in litter ( $r^2=0.92$ ). Soil N levels alone explain over 90% of Hg pool sizes across the four Sierra Nevada forest sites. This suggests that soil organic N and C groups provide sorption sites for Hg to retain atmospheric deposition. However, the patterns could be due to

indirect relationships where high soil N and C levels reflect high ecosystem productivity which leads to corresponding high atmospheric Hg deposition inputs via leaf litterfall and plant senescence. Our results also show that two of the sites previously affected by prescribed burning and wildfires show significant depletion of above-ground Hg pools but that belowground Hg pools remain unaffected. We conclude that sequestration of Hg in remote Sierra Nevada forest sites is strongly co-determined by ecosystem parameters with C and N pools being excellent determinants for the pool sizes of Hg.

## 1 Introduction

Significant quantities of mercury (Hg) primarily derived from the atmosphere are associated with vegetation in terrestrial ecosystems, and large pools of atmospheric Hg deposition is retained in surface litter and soil pools (Mierle, 1990; Grigal, 2003; Munthe et al., 1998). Hg pools in terrestrial ecosystems greatly exceed annual fluxes to and from these systems (Mason et al., 1994; Grigal et al., 2000; Schwesig and Matzner, 2000; Lindberg, 1996; Munthe et al., 1998). Retention and sequestration of Hg in vegetation and surface soils is attributed to a strong affinity of Hg to organic matter (OM; Meili, 1991; Yin et al., 1996; Schuster, 1991). Since terrestrial ecosystems are the main source of Hg to many aquatic ecosystems (e.g., Lorey and Driscoll, 1999; Swain et al., 1992), accumulation and retention of Hg in terrestrial compartments and the mobility of Hg within ecosystems are key to understand how atmospheric Hg loadings will ultimately affect aquatic ecosystems where fish Hg can build up



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to harmful levels even in relatively remote areas such as the Western United States (Peterson et al., 2007).

There is strong evidence that most Hg allocated in above-ground plant compartments is of atmospheric origin and that above-ground plant Hg uptake hence represents an important pathway for atmospheric deposition (e.g. Iverfeldt, 1991; Munthe et al., 1995; Rea et al., 1996; Lindberg, 1996). Plant and ecosystem uptake of atmospheric Hg may thereby feed back on atmospheric Hg levels (Obrist, 2007; Obrist et al., 2008a). Evidence for the atmospheric origin of above-ground tissue Hg is based on limited translocation of Hg within plants (Beauford et al., 1977; Godbold and Hüttermann, 1988; Lindberg et al., 1979; Bishop et al., 1998; Schwesig and Krebs, 2003); measured assimilation of airborne Hg by stomatal uptake at the plant level (Hanson et al., 1995; Ericksen et al., 2003) and at the ecosystem level (Obrist et al., 2006; Fritsche et al., 2008a); and combined air/soil exposure experiments (Ericksen et al., 2003; Frescholz et al., 2003; Millhollen et al., 2006a, b). Hg concentration and accumulation in vegetation, litter, and soils of terrestrial ecosystems show some typical patterns as reported for forests (review by Grigal, 2003): (i) Tree foliage show a mean concentration of  $26 \mu\text{g kg}^{-1}$  with most tree foliage Hg levels ranging between 10 to  $100 \mu\text{g kg}^{-1}$ , and concentrations generally increase with foliage age (Rasmussen, 1995). (ii) Bole wood – which accounts for up to 75% of above-ground biomass (Johnson and Lindberg, 1992) – shows low Hg concentrations, typically between 1 to  $4 \mu\text{g kg}^{-1}$ , but significantly higher concentrations ( $>1$  order of magnitude) are usually found in tree bark; (iii) Forest floor concentrations (i.e., litter horizons) in North-Central United States average around  $140 \mu\text{g kg}^{-1}$  and are typically higher than concentrations of the underlying mineral soils; and (iv) close relationships are observed between litter and soil Hg and organic matter contents, but the stoichiometry (e.g. Hg/C ratios) can vary widely across different sites, soils, and soil depths.

Understanding what factors determine plant, litter, and soil Hg contents is important to predict the fate and stability of atmospherically derived Hg in terrestrial ecosystems with important implications for the loading of Hg to water bodies. Comparisons across different sites are challenging because of different site histories, different measurements protocols, and regionally variable atmospheric deposition loads. In fact, transect and spatial studies are used to infer the influence of local and regional pollution on Hg levels in litter, humus, and surface soils (Nater and Grigal, 1992; Håkanson et al., 1990; Låg and Steinnes, 1978). This study aims to compare Hg concentrations and pools sequestered in major ecosystem compartments and their relationships to the respective carbon (C) and nitrogen (N) contents in four remote forest sites in the Sierra Nevada mountain range of the Western United States, as part of a larger project aimed at quantifying Hg pools and fluxes across US forest ecosystems (EPA STAR #RD833378010). All sites are in close proximity with similar precipitation regimes, elevations, and climatic con-

ditions, and hence likely experience similar atmospheric deposition loads. This presents a unique opportunity to assess how ecosystem parameters (e.g. species, ecosystem productivity, biomass and litter accumulation) and soil properties (soil texture, soil C and N contents) control accumulation and retention of Hg in terrestrial ecosystems. In addition, two sites each are collocated within 300 m distance on the same parental materials with one site each having experienced recent forest floor and/or vegetation removal by a wildfire or prescribed burning, which allows to evaluate the dependence of ecosystem Hg pools with respect to fire history. We present detailed stoichiometric relations of Hg with C, N contents and C/N ratios to evaluate how relations change as Hg moves through ecosystem compartments (e.g. from leaves to soil organic C pools) and to infer about causes and fate of Hg sequestration in terrestrial ecosystems.

## 2 Materials and methods

### 2.1 Study sites and locations

All four study sites are located on the east lee-ward side of the Sierra Nevada near Lake Tahoe at the border between California (CA) and Nevada (NV) at elevations between 1700 to 2010 m a.s.l. Two sites each are co-located within very short distances (300 m). Two sites are located ca. 32 km north of Truckee, CA (termed “Truckee, CA”) at an elevation of 1770 m a.s.l. Overstory vegetation is dominated by Jeffrey pine (*Pinus jeffreyi* Grev. and Balf.) and a few scattered White Fir (*Abies concolor* (Gord. and Glend.) Lindl.) with little understory vegetation. Soils are the Kyburz series, fine-loamy mixed, frigid Ultic Haploxeralfs derived from andesite. A close weather station near the Truckee site lists yearly precipitation of 568 mm (Boca, elevation: 1700 m a.s.l.; [www.wrcc.dri.edu](http://www.wrcc.dri.edu)). Two further sites are located 30 km southwest of Reno at an elevation of 2010 m and are termed “Little Valley, NV”. The sites experience average annual precipitation of 550 mm. Vegetation at the site is dominated by Jeffrey pines (*Pinus jeffreyi* Grev. and Balf.), and soil are the Corbett series, sandy and fine-loamy mixed, frigid Typic Xeropsamments derived from colluvium of decomposed granite. The distance between the two pairs of adjacent sites is approximately 50 km in a straight line.

Much of the Eastern Sierra Nevada Mountains were logged and probably burned during the Comstock mining era between 1870 and 1890, so typical tree age of the dominant pine forests ranges between 120 and 140 years. In one of the two Truckee sites (called post-fire site), a prescribed burning treatment in 2002 resulted in a loss of 62% of the forest floor mass (Murphy et al., 2006), but the fire did not remove any of the overstory trees. No prescribed burning was conducted in an adjacent site called intact. In Little Valley, a stand-replacing wildfire in 1981 occurred in the “post-fire” site, which was followed by salvage logging in the following

year to remove all merchantable timber. An adjacent “intact” site received some under-burning (i.e. forest floor burning, but not crown fire) with no mature tree mortality. The post-fire area has been re-vegetated by a shrub community now dominated by Manzanita (*Arcostaphylos patula* Greene) and snowbush (*Ceanothus velutinus* Dougl.), a species that readily invades after fire and is a N fixing species (Youngberg and Wollum 1976).

## 2.2 Sample collection for measurement of Hg, C, and N mass ratios and soil properties

Field samples for quantification of Hg contents and pools were collected in October and November 2007 in all four sites. We followed similar field sampling protocols (e.g. same sampling plots, replications, etc.) as were used in previous studies for biomass, C, and N stock quantification (Johnson et al., 1997, 2001, 2005; Murphy et al., 2006) to be able to scale up Hg concentrations to ecosystem-level pools (see below). Samples were taken in 4 replicate plots in each site (with the exception of 3 replicate plots in the Little Valley intact site), and 3 random locations within each replicate plot were sampled for one pooled sample per plot; i.e. 12 (and 9) sampling locations per site. Plot sizes were 0.04 ha and 0.1 ha, respectively, with approximate distance of 50 to 100 m between plots. Ecosystem compartments sampled included green leaves/needles, dry leaves/needles, branches, bark, bole wood, understory vegetation, litter horizons (Oi: undecomposed surface litter, Oe: partially decomposed litter, Oa: decomposed organic humus, and coarse woody debris), and different soil layers. All sampling was conducted using clean latex gloves and sampling equipment made of stainless steel, and all sampling equipment was washed prior to use in the field using de-ionized water. All samples were immediately double-bagged in plastic freezer bags and were stored on ice until transport to the laboratory and transfer to a  $-25^{\circ}\text{C}$  freezer on the same day. Green and dry needles and leaves and branches were directly cut or picked from trees and shrubs using scissors and clippers, bark was removed by cutting using a knife, and bole wood was cored (after removal of the surrounding bark) using a 40 cm long, 5.15-mm core diameter tree borer. Forest surface litter (Oi, Oe, Oa, and coarse woody debris) was picked directly from the floor using clean gloves and a spade. Soils at different depths (0–20, 20–40 for the Truckee sites, 0–7, 7–20, and 20–40 for the Little Valley sites) were sampled using stainless-steel hand-shovels from the walls of larger soil pits excavated with shovels and pick axes.

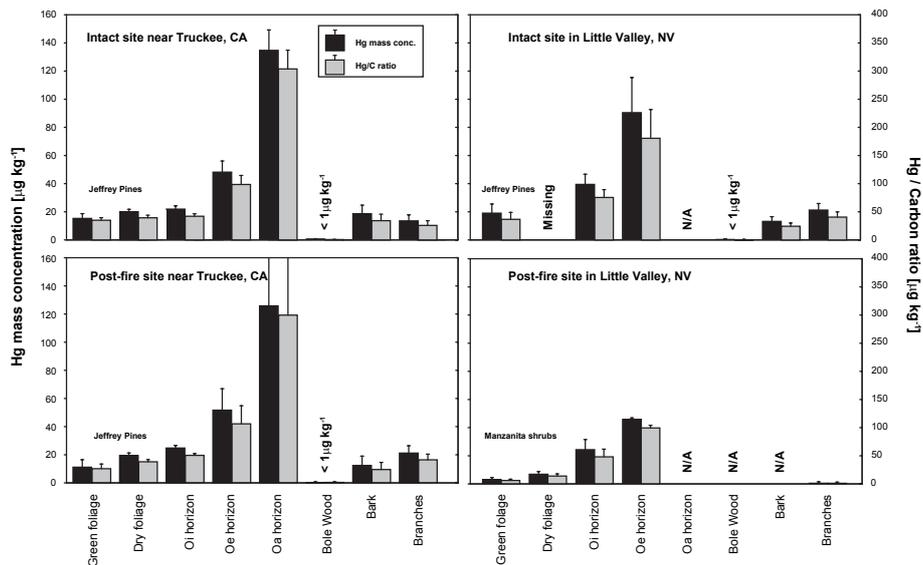
## 2.3 Tissue, litter, and soil sample preparations and sample analyses

All samples were freeze dried in acid-cleaned (5%  $\text{HNO}_3$ ) 25-ml volume test tubes using a freeze dryer with an 8-port column flask (1-l volumes) manifold and a  $-50^{\circ}\text{C}$  stain-

less steel ice condenser (Model MicroModulyo-115, Thermo Scientific, Waltham, MA, USA). After freeze drying, samples were milled and homogenized using stainless steel coffee mills. Samples were analyzed for total Hg concentrations using a Nippon Model MA-2000 total mercury analyzer in accordance with US EPA Method 7473. The analyzer was calibrated using 0.1 ppm and 0.01 ppm Hg stock made of a 1000 ppm  $\text{HgCl}_2$  standard (FGS chemicals) in 0.001% L-cysteine solution according to the manufacturer’s manual. National Institute of Standards and Technology (NIST) solid standard reference materials (#1515: Apple leaves;  $44 \pm 4 \mu\text{g Hg kg}^{-1}$ ) were run after every 8 samples. When NIST standards deviated  $>5\%$  from the expected values, the analyzer was recalibrated immediately and all samples since the last acceptable NIST standard were re-run. The 124 performed NIST standards showed excellent data quality with an average concentration value of  $44.6 \pm 1.6 \mu\text{g kg}^{-1}$  (mean  $\pm$  standard deviation) and a coefficient of variability (CV) of 3.6%. Samples were analyzed in duplicates and analyses repeated when the CV of two samples exceeded 10%, with the exception of concentration values below 10 ng/g where a sample CV of 20% was allowed. Total C and N concentrations and C/N ratios were analyzed using a Leco Truspec carbon/nitrogen analyzer (LECO, St. Joseph, Michigan, USA) at the Soil Forage and Water Analysis Laboratory at Oklahoma State University (<http://www.soiltesting.okstate.edu/>). Soil texture was analyzed at the same laboratory using the hydrometer method which determines percent sand, silt, and clay settling rates as measured by a ASTM 152-Type hydrometer. Corrections for viscosity and temperature were made by use of an analytical blank and by adjusting the time for the final hydrometer reading based on temperature tables. The laboratory guarantees accuracy and precision of test results through daily analysis of quality control samples, a three step internal data review process, and participation in external certification and sample exchange programs. All instruments are calibrated with certified standards and maintained according to the specification. Specific quality control for C/N analysis included frequent blank test and sample checks using standards every 10 samples, and one blank and one check sample for each of analyses for soil texture analysis.

## 2.4 Scaling of measured Hg and C concentrations to the ecosystem level to calculate above- and below-ground Hg pools

For scaling measured Hg concentrations and Hg/C ratios to the level of ecosystems, we used previous data from studies with detailed quantification of above- and belowground biomass and nutrient pools (Johnson et al., 2005; Murphy et al., 2006). In the two adjacent Truckee sites, replicate circular plots of 0.04 ha in size were established in the intact and post-fire sites and four replicate plots each were sampled in 2001 and 2002 for biomass and litter dry mass



**Fig. 1.** Concentrations of Hg (black bars; left axis) and Hg/C ratios (gray bars; right axis) of the main ecosystem components of four Eastern Sierra Nevada sites. Ecosystem components include green and dry foliage, wood, bark and branches of the dominant trees and shrubs per site and the surface litter horizons (Oi: undecomposed surface litter; Oe: partially decomposed litter; Oa: decomposed organic humus).

estimation (per unit area) and for determination of soil properties (Johnson et al., 2008; Murphy et al., 2006). Tree diameter were measured at breast height (dbh) for all trees in all sampling plots and dry mass of foliage, branches, bole wood, and bole bark were estimated for each plot using established regressions of tree diameter and the respective biomass components (Gholz et al., 1979; Johnson et al., 2008). Forest floor (i.e. litter) biomass was destructively sampled in 2002 (post-fire plots) and 2001 (intact plots) in six randomly established 15-cm diameter litter rings per plot with the following categories: woody material up to 2.54 cm diameter, Oi, Oe and Oa (Murphy et al., 2006). Quantitative litter samples were sieved (0.84 mm standard testing sieved) to separate any mineral fraction from the organic fractions, and samples were dried to calculate area averaged forest floor biomass ( $\text{kg ha}^{-1}$ ). After removal of the litter cover, soils at each point were sampled by depth (0–20 cm, 20–40 cm, and 40–60 cm) using a bucket auger. In order to estimate soil mass, quantitative pits were dug near the center of each plot and all soil and rock material was weighed (Johnson et al., 2008). Soil samples were oven dried at  $55^{\circ}\text{C}$  until stable weight for calculation of bulk density to calculate area-based soil mass. In the Little Valley sites, replicate plots of 0.1 ha were established both in the intact (three plots) and post-fire site (four plots) with similar slopes and exposures (Johnson et al., 2005). Two sites (post-fire) and one site (intact) were sampled for biomass and soil mass in 1997 while two more sites each were sampled in 2001. In the intact site, all trees were measured at dbh for estimation of biomass of all major tree components (i.e. leaves, branches, bole, bark; see above). In the post-fire shrub plots, six randomly es-

tablished  $1\text{ m}^2$  subplots were destructively harvested for all aboveground biomass by cutting all aboveground vegetation (sorted by species and component – foliage and woody) for estimation of dry mass. Litter (by horizons) was sampled with 15 cm diameter litter rings, and soils were removed (0–7 cm, 7–2 cm, 20–40 cm) and soil mass estimated for each horizon were measured by the quantitative pit method (Johnson et al., 2005).

By using estimated biomass pools from these previous studies, calculated ecosystem-levels of Hg represent pools that were present at the sites in the years 2001/2002 and 1997/2001, respectively. Since annual fluxes (deposition, runoff, emissions) generally represent only a minor fraction of available ecosystem pools (Mason et al., 1994; Grigal et al., 2000; Schwesig and Matzner, 2000), we expect that current ecosystem-level Hg pools would have changed only slightly. Similarly, seasonal differences in Hg contents may result in small differences in stocks throughout the year, but these are considered small compared to the large stocks present in these ecosystems.

### 3 Results and discussion

#### 3.1 Above-ground Hg concentrations

Figure 1 shows Hg concentrations and Hg/C ratios of the main tree and shrub species of the four sites, and respective values are also listed in Table 1 (means and standard errors of replicate sampling plots per site). Hg concentrations of green needles in Jeffrey Pine, the dominant tree in all forest sites, range between  $11.2$  and  $18.9\ \mu\text{g kg}^{-1}$ . Concentrations

**Table 1.** Hg concentrations, Hg/C and Hg/N ratios of foliage, litter, and soils.

		Truckee		Little Valley	
		Intact	Post-fire	Intact	Post-fire
A: Hg concentrations ( $\mu\text{g Hg/kg}$ dry mass) of ecosystem components in four remote Sierra Nevada forest sites.					
Foliage	Green foliage	J. Pine: 15.0 $\pm$ 1.5 Fir: 49.2 $\pm$ 2.3	J. Pine: 11.2 $\pm$ 2.4	J. Pine: 18.9 $\pm$ 3.8	Manzanita: 2.9 $\pm$ 0.8 Ceonothus: 6.1 $\pm$ 0.7 J. Pine Seedling: 9.4 $\pm$ 1.2 Manzanita: 6.8 $\pm$ 0.10
	Dry foliage	J. Pine: 19.7 $\pm$ 0.8 Fir: 57.0 $\pm$ 4.7	J. Pine: 19.5 $\pm$ 0.9	N/A	
Litter	Oi	21.6 $\pm$ 1.0	24.9 $\pm$ 0.8	39.4 $\pm$ 4.2	24.3 $\pm$ 3.6
	Oe	47.7 $\pm$ 3.5	51.9 $\pm$ 6.8	90.5 $\pm$ 14.4	45.9 $\pm$ 0.6
	Oa	133.7 $\pm$ 6.5	126.4 $\pm$ 21.4	N/A	
Soils	0–7 cm	35.3 $\pm$ 3.1	29.9 $\pm$ 1.9	12.8 $\pm$ 0.4	12.5 $\pm$ 0.9
	7–20 cm	23.6 $\pm$ 0.8	25.0 $\pm$ 0.6	8.5 $\pm$ 1.4	9.7 $\pm$ 0.7
	20–40 cm			7.5 $\pm$ 0.5	7.9 $\pm$ 0.5
B: Hg/C ratios ( $\mu\text{g Hg/kg C}$ ) of same components					
Foliage	Green foliage	J. Pine: 34 $\pm$ 2 Fir: 95 $\pm$ 6	J. Pine: 25 $\pm$ 4	J. Pine: 37.7 $\pm$ 7	Manzanita: 2.9 $\pm$ 1 Ceonothus: 12 $\pm$ 1 J. Pine Seedling: 18 $\pm$ 2 Manzanita: 14 $\pm$ 2
	Dry foliage	J. Pine: 39 $\pm$ 2 Fir: 110 $\pm$ 9	J. Pine: 37 $\pm$ 2	N/A	
Litter	Oi	42 $\pm$ 2	49 $\pm$ 1	76 $\pm$ 8	48 $\pm$ 7
	Oe	98 $\pm$ 7	105 $\pm$ 14	181 $\pm$ 30	99 $\pm$ 2
	Oa	304 $\pm$ 15	298 $\pm$ 50	N/A	
Soils	0–7 cm	762 $\pm$ 102	576 $\pm$ 54	871 $\pm$ 104	764 $\pm$ 152
	7–20 cm	759 $\pm$ 110	734 $\pm$ 70	1098 $\pm$ 139	764 $\pm$ 101
	20–40 cm			982 $\pm$ 75	948 $\pm$ 114
C: Hg/N ratios (mg Hg/kg N) of the same components					
Foliage	Green foliage	J. Pine: 1.7 $\pm$ 0.1 Fir: 6.4 $\pm$ 0.4	J. Pine: 1.2 $\pm$ 0.2	J. Pine: 1.8 $\pm$ 0.5	Manzanita: 0.3 $\pm$ 0.1 Ceonothus: 0.5 $\pm$ 0.1 J. Pine Seedling: 0.9 $\pm$ 0.1 Manzanita: 0.8 $\pm$ 0.1
	Dry foliage	J. Pine: 4.5 $\pm$ 0.1 Fir: 14.2 $\pm$ 0.8	J. Pine: 4.6 $\pm$ 0.3	N/A	
Litter	Oi	4.7 $\pm$ 0.4	5.2 $\pm$ 0.2	8.9 $\pm$ 0.2	3.3 $\pm$ 0.8
	Oe	5.7 $\pm$ 0.4	5.9 $\pm$ 0.8	10.0 $\pm$ 1.0	4.8 $\pm$ 0.2
	Oa	11.6 $\pm$ 1.2	9.2 $\pm$ 0.6	N/A	
Soils	0–7 cm	19.0 $\pm$ 1.6	15.3 $\pm$ 1.1	19.2 $\pm$ 0.9	19.5 $\pm$ 2.7
	7–20 cm	18.4 $\pm$ 0.8	17.3 $\pm$ 1.1	21.9 $\pm$ 2.9	18.4 $\pm$ 3.2
	20–40 cm			21.3 $\pm$ 3.6	21.3 $\pm$ 2.5

of Hg in White Fir, which is occasionally present in the Truckee sites, is considerably higher with an average concentration of 49.2  $\mu\text{g kg}^{-1}$ . In the post-fire Little Valley site, which is devoid of trees, the dominant shrub leaf concentrations and concentration of emerging pine seedlings are lower (2.9  $\mu\text{g kg}^{-1}$  in Manzanita shrubs, 6.1 $\pm$ 0.7  $\mu\text{g kg}^{-1}$  in Ceonothus shrubs, and 9.4  $\mu\text{g kg}^{-1}$  in pine seedlings). Of other above-ground biomass tissues, branches and bark exhibit significant Hg concentrations (12.4–21.3  $\mu\text{g kg}^{-1}$ ), while bole wood concentrations across all sites were much

lower (0.23 $\pm$ 0.12  $\mu\text{g kg}^{-1}$  for Jeffrey Pine and below the detection limit in White Fir). Manzanita shrub branches also showed very low levels of Hg (0.37 $\pm$ 0.51  $\mu\text{g kg}^{-1}$ ).

Our measured foliar Hg levels generally agree well with published concentrations: Grigal (2003) lists a midpoint Hg concentration of 24  $\mu\text{g kg}^{-1}$  for live foliage, and a series of papers list foliage concentrations of various tree species between 15 and 51  $\mu\text{g kg}^{-1}$  (St. Louis et al., 2001; Grigal et al., 2000; Rasmussen, 1995; Moore et al., 1995; Zhang et al., 1995). Bole wood concentrations measured at our sites,

however, are lower than some of the data reported in the literature. Grigal (2003) reviewed a series of studies reporting bole wood concentrations between 2 to 13  $\mu\text{g kg}^{-1}$  with concentrations in the north-central part of the USA ranging from 0.6 to 12  $\mu\text{g kg}^{-1}$  (with 60% of the values in the ranging between 1 to 4  $\mu\text{g kg}^{-1}$ ). Engle et al. (2006) report bole wood concentrations in the same Truckee site as this study of 2.9  $\mu\text{g kg}^{-1}$ . The lower concentrations measured in our study could be due to the remote location of the Sierra Nevada. A second reason for the difference could be that we carefully removed the surrounding bark before coring trees, and if other studies were to include any bark while coring trees, they would show higher levels as bark shows considerably larger Hg levels than the bole wood (Table 1, Fig. 1).

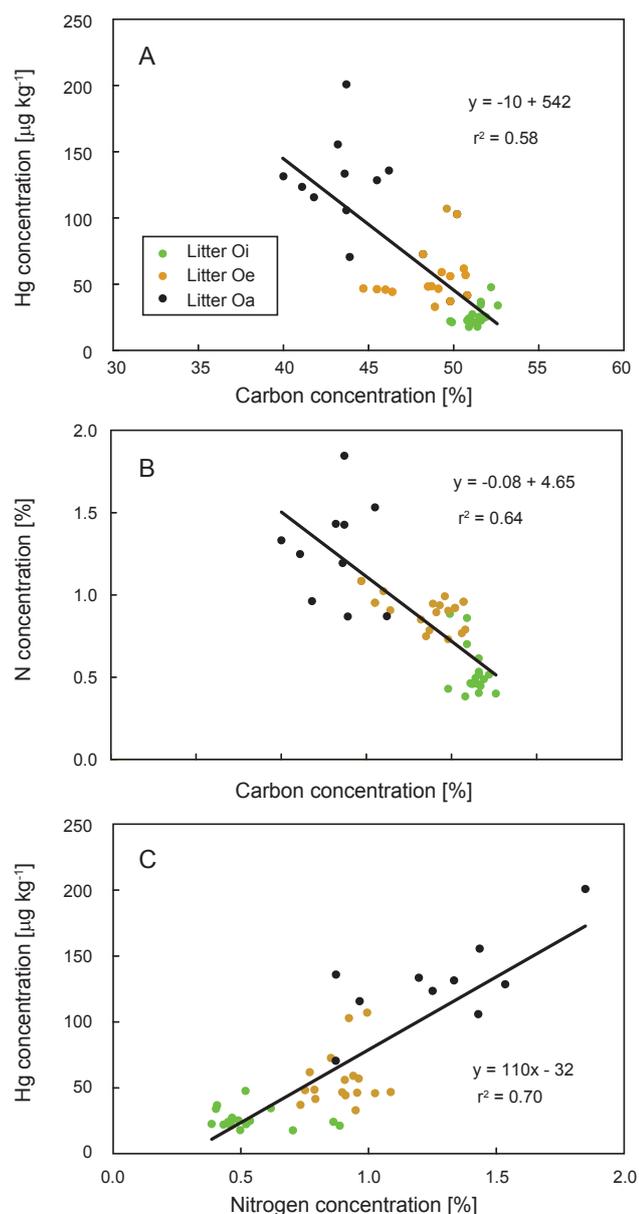
### 3.2 Trends in leaf and litter Hg concentrations and stoichiometry in respect to C and N

Our study highlights important trends in concentrations from green leaves and needles to dry foliage and litter horizons, which were mainly composed of fallen and decomposing needles and leaves of the dominant trees and shrubs since understory vegetation was sparse. Hg concentrations and Hg/C ratios increase in the following order: Green Needles/Leaves < Dry Needles/Leaves < Oi horizon (undecomposed surface litter) < Oe horizon (partially decomposed litter) < Oa (decomposed organic humus). It has previously been reported that leaves and needles in litterfall have higher concentrations than those measured in green foliage (40–80% higher; Grigal et al., 2000), an effect likely due to increasing foliar Hg concentrations with increasing leaf age (Rasmussen, 1995; Fleck et al., 1999; Frescholtz et al., 2003; Milhollen et al., 2006a, b; Munthe et al., 1998). Hg concentrations of green and dry needles and leaves directly picked from trees and shrubs in this study support this finding with dry foliage concentrations exceeding green foliage Hg levels by 16% (Fir), 31 to 74% (Jeffrey Pine), and even 134% (Manzanita).

Hg concentrations in the litter horizons increase from the initial Oi litter horizon to successively decomposing litter horizons Oe and Oa, with Oe horizons in all sites showing roughly double the Hg contents of the initial Oi horizons (Table 1). In the Truckee sites, the only sites with developed Oa horizons, these humus layers show Hg concentration 4–5 times higher than the levels measured in the initial Oi litter horizons. Increased Hg concentrations in litter is accompanied by an even stronger change in stoichiometric relations. Hg/C ratios increase by a factor of almost 100 from green foliage to the well decomposed Oa litter horizons (Table 1, Fig. 2). Grigal et al. (2000) reported Hg concentration increases in a hardwood forest in Minnesota from 21  $\mu\text{g kg}^{-1}$  in litterfall foliage to 84  $\mu\text{g kg}^{-1}$  in the forest floor. Similar, Friedli et al. (2007) reported surface litter levels between 68 and 127  $\mu\text{g kg}^{-1}$  in a boreal forest site in Canada and higher Hg contents in organic layers (100 to 250  $\mu\text{g kg}^{-1}$ ),

and Åkerblom et al. (2008) reported higher Hg/C ratios in the lower half of forest mor layers as compared to the upper half in boreal forest sites. Hall and St. Louis (2004) reported total Hg mass increases in decomposing boreal forest leaves and needles during an 800 day in situ decomposition study using litter bags along with decreases in C and N. Åkerblom et al. (2008) attributed this trend to a natural downward enrichment of Hg due to soil decomposition or other processes, and Grigal (2003) similarly concluded that a higher Hg density of organic matter in the mineral soil as compared to the forest floor implies that less Hg than C is lost during OM mineralization. Similar enrichment of Hg in plant detritus was first reported for aquatic ecosystems by Lindberg and Harriss (1974).

Figure 2a shows declining C concentrations and increasing Hg concentrations from Oi to Oe and Oa litter horizons, and the pattern for N in litter is almost identical to that for Hg (Fig. 2b). Litter N is known to be fairly well retained during decomposition processes hence leading to decreased C/N ratios as litter decomposes. Hence, these patterns may support Åkerblom's and Grigal's notion that during decomposition C is lost as  $\text{CO}_2$  while Hg – as well as N – are efficiently retained in the remaining decomposing litter and soil substrate. However, it is important to keep in mind that other processes also may contribute to this trend. For example, the three litter horizons have very different ages and therefore have been exposed to environmental influences for very different time spans. Soil organic C can be as old as 6000 years, (Eusterheus et al., 2006), and a reasonable estimate of the age of the Oa litter in Sierra Nevada forest sites of this study may be around 110 to 130 years, which is approximately the stand age and time since the last major fire (Johnson et al., 2005). Initial surface Oi litter samples on our sites clearly originated from the current year of foliage. Hence, accumulation of Hg in decomposing litter does not necessarily mean preferential retention of Hg over C during decomposition, but could simply be due to continued atmospheric deposition inputs of Hg to the older decomposing litter. Soil organic matter in fact shows a strong affinity for oxidized Hg in solution (Yin et al., 1996) and hence for atmospheric Hg deposition. We propose that the observed increase in Hg in decomposing litter horizons may not solely be driven by retention of Hg during losses of C but are also due to retention of continued atmospheric deposition. If, for example, an assumed annual atmospheric deposition load of 20  $\mu\text{g m}^{-2}$  were to be deposited and fully retained by the litter, this would lead to concentration increases in litter of  $\sim 9 \mu\text{g kg}^{-1}$  per year (using the average total litter mass of 2.165  $\text{kg m}^{-2}$  present at the sites). This is a signal strong enough to lead to significant concentration increases over the time spans of the litter. In our view, experimental studies (cf. Fritsche et al., 2008b) are needed to clarify the fate of Hg in decomposing organic material and to clarify how atmospheric deposition versus loss of Hg to solution or back to the atmosphere may contribute to Hg enrichment in decomposing organic material. This has large



**Fig. 2.** Scatter plots and linear regressions between (a) litter Hg and C concentrations; (b) litter N and C concentrations, and (c) litter Hg and N concentrations of the four sites. Green symbols represent Oi litter (undecomposed surface litter), red symbols represent Oe (partially decomposed litter), and black symbols Oa litter samples (decomposed organic humus). All linear regressions are statistically significant ( $P < 0.01$ ).

implications for the fate of mercury stored in global organic C pools, especially in light of future climate change effects on global C pools, and may possibly feed back on global atmospheric Hg levels (cf. Obrist, 2007).

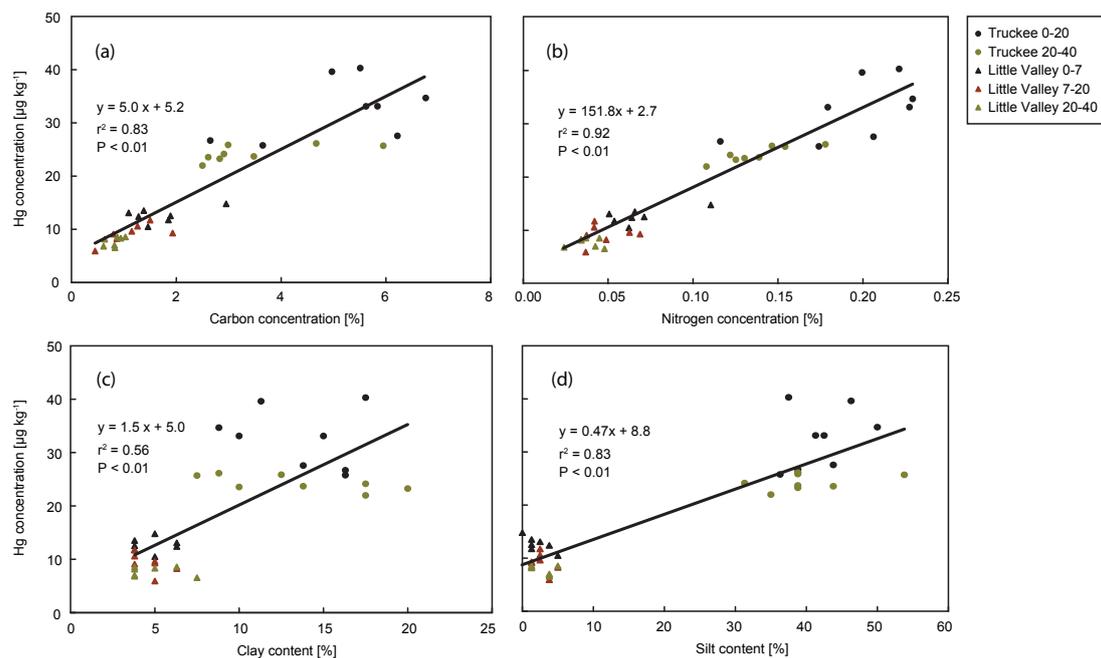
The resulting inverse relationship between Hg and C observed across the various litter horizons (Fig. 2; slope:

$-1 \mu\text{g g}^{-1}$ ;  $r^2=0.58$ ) is in contrast to reported positive relationships between Hg and C (Aastrup et al., 1991; Meili, 1991; Grigal, 2003) in forest floors, humus layers, and soils (see also discussion below). To our knowledge, however, no study has yet compared in detail at Hg/C relationships in differently decomposed litter horizons. Our results indicate that Hg and Hg/C concentrations in pure organic layers such as litter depend on the decomposition stage and may not follow the general pattern observed in mixed organic layers (e.g. mineral soils or mor layers) where Hg is positively related to organic matter contents (see soil results below). Figure 2 shows that Hg levels are positively correlated to litter N levels, with similar slopes ( $11 \text{ g g}^{-1}$ ;  $r^2=0.70$ ) to a strong linear relationship between Hg and N observed in mineral soil horizons (see Fig. 3). This also suggests that organic N groups in litter and soils may be important sorption sites in terrestrial ecosystems (see further discussion below).

### 3.3 Soil Hg concentrations and relationship to soil C and N

Soil Hg concentrations range between  $7.5$  to  $35.3 \mu\text{g kg}^{-1}$  across all sites (Fig. 3, Table 1) and are hence substantially lower than in the overlying litter horizons. Deeper soil layers show lower Hg concentrations as compared to upper soil horizons which is evident across all sites. The Truckee sites show 16 to 33% lower Hg concentrations in the lower half (20–40 cm) of the sampled soil profile. A more detailed depth sampling in the Little Valley sites (0–7 cm, 7–20 cm and 20–40 cm) indicates that the strongest concentration drop occurs near the very top soil layers. Declining Hg concentrations with soil depth are commonly observed: Aastrup et al. (1991) reported declining Hg concentrations from mor horizons to B and C horizons (from  $250$  to  $6 \mu\text{g kg}^{-1}$ ) in a pine/spruce forest in Sweden; soil concentrations decreased sharply with depth in mineral horizons in coniferous and deciduous forests in Germany (Schwesig and Matzner, 2000); sites in central Minnesota showed marked declines from the top 0–10 cm to 10–50 cm depth (from  $36$  to  $11 \mu\text{g kg}^{-1}$ ; Grigal et al., 1994); and Engle et al. (2006) reported decreasing Hg concentrations in two Sierra Nevada forest sites with highest Hg concentrations in the top A horizons. Exceptions to these patterns have been reported in podzolic and chernozemic soils where leaching and accumulation horizons of organic matter and sesqui-oxides are believed to cause different depth distribution of Hg (Schwesig and Matzner, 2000; Dudas and Pawluk, 1976).

The depth patterns of Hg in terrestrial soils are generally attributed to a close relationship between Hg and soil organic matter (Aastrup et al., 1991; Meili, 1991; Grigal, 2003), which usually shows pronounced concentration declines with depth (Johnson and Lindberg, 1992). This relationship is also obvious in our study where data show positive linear regressions ( $P < 0.01$ ) between organic C and soil Hg concentrations across all sites and soil horizons



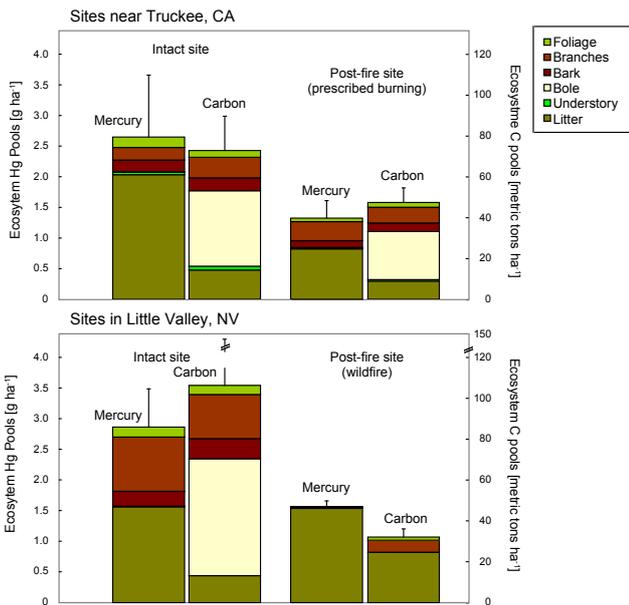
**Fig. 3.** Scatter plots and linear regressions between (a): soil Hg and C concentrations; (b) soil Hg and N concentrations; (c) soil Hg and clay content; and (d) soil Hg and silt content. Circles represent the two sites near Truckee, CA and triangles represent the Little Valley, NV sites (no significant differences between the intact and post-fire site). Black symbols are for top soils (0–20 cm in Truckee, 0–7 cm in Little Valley); brown symbols for middle soil layers (7–20 cm in Little Valley), and green symbols are for the lowest measured soil layers (20–40 cm at all sites).

(Fig. 3). In this figure, each data point represents an individual plot-level measurement (4 and 3 plots per site, respectively) at different soil depths, with adjacent intact and post-fire sites not separated visually due to a lack of fire effects on belowground Hg pools and patterns (see below). A combined linear regression across all sites and horizons shows a slope of  $0.5 \mu\text{g Hg g}^{-1} \text{C}$  ( $P < 0.01$ ;  $r^2 = 0.83$ ). A review by Grigal (2003) reports close relationships between Hg and C across various sites showing similar changes in Hg concentration per unit change in organic matter (about  $0.22 \mu\text{g Hg per g soil organic matter, SOM}$ ). Using a C/SOM mass ratio of  $\sim 0.5$  measured on our sites, the calculated slope in our study (i.e.  $0.25 \mu\text{g Hg per g SOM}$ ) is close to the ratio reported by Grigal (2003). Låg and Steinnes (1978) report a clear correlation between Hg and organic matter content in humus layers of Norwegian forest soils, but their reported slopes – as well as total humus Hg levels – are much lower as compared to other studies. Munthe et al. (1998) summarize results from catchments in Sweden from the surface to 70 cm depths showing a close association between ignition losses (a measure for organic content in the soil) and both total and methylated Hg.

An even stronger linear relationship, however, was observed in our data between soil Hg and soil N. This relationship was significant in all sites individually, with no statistically significant difference in slopes and intercepts between

the sites. A combined linear regression showed a slope of  $15.2 \mu\text{g Hg g}^{-1} \text{N}$  and indicates that soil N contents alone explained a full 92% of the Hg variability observed across all sites and soil horizons. The affinity of Hg to organic matter has been attributed to complexation of Hg with reduced S and O or N groups abundantly present in organic molecules (Schuster, 1991; Skyllberg et al., 2000; Xia et al., 1999). Our results showing strong relationships between Hg and N in soils and similar relationships in litter (see above) suggest that nitrogen groups may be key ligands for retention of Hg in terrestrial ecosystem compartments.

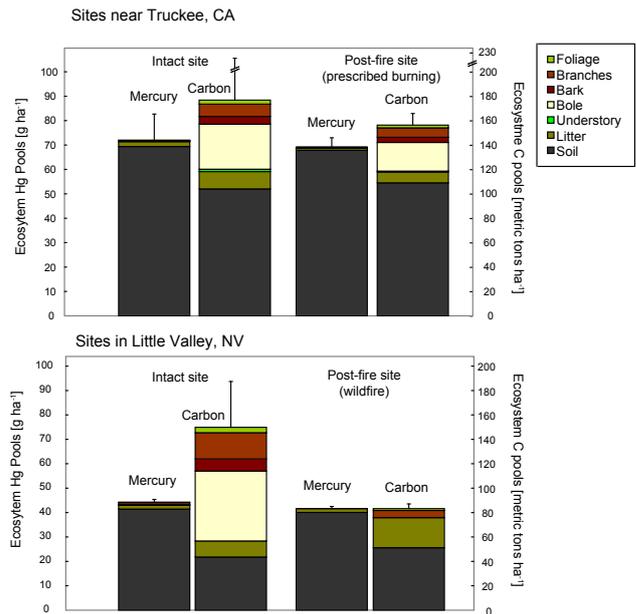
Soil texture showed no relationships to Hg contents within any individual site, but there was a relationship to clay and silt content across all sites. The relationships, however, were weak and caused by the Truckee sites with higher silt and clay contents also showing higher Hg concentrations as compared to the sandier Little Valley sites. The lack of correlation of Hg content with clay content within individual sites, however, was surprising since a recent review (Gabriel and Williamson 2004) about Hg in soils and terrestrial environment suggested that clay minerals – along with a series of oxides and metal hydroxides – play an important role as sorbent for Hg in soils. However, differences in soil texture between the Truckee and Little Valley sites may co-determine differences in retention of C, N, and Hg at the sites.



**Fig. 4.** Total ecosystem above-ground pools of Hg (left bars; left axis) and corresponding C pools (right bars, right axis) in four Sierra Nevada sites. Error bars represent standard error of pools calculated for individual 4 (3 for Little Valley intact site) replicate plots per site.

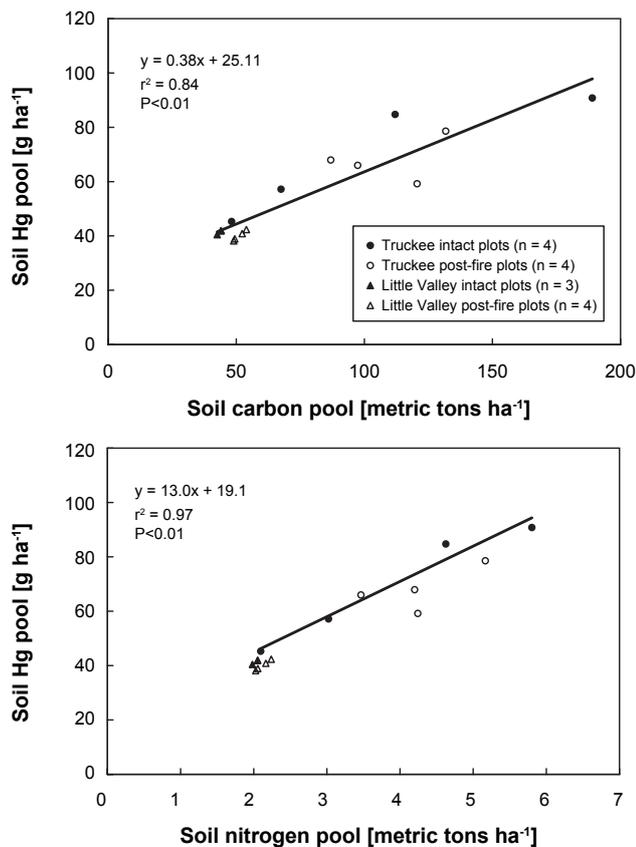
### 3.4 Ecosystem-level pools of Hg and C

Figure 4 shows aboveground Hg and C pools of the four sites scaled up to the ecosystem levels. In the intact sites, above-ground Hg pools contain  $2.65 \pm 1.01 \text{ g Hg ha}^{-1}$  near Truckee and  $2.87 \pm 0.62 \text{ g Hg ha}^{-1}$  in Little Valley. In both sites, the largest Hg pools are located in the litter horizons which accounted for more than 75% in the Truckee sites and for just over 50% in the Little Valley sites. The second largest pools are measured in branches (11% to 23% of above-ground Hg) and further significant Hg pools can be found in bark and needle biomass, two pools which likely originate from sorbed deposition of atmospheric origin. Although bole wood presents a major fraction of C allocation in both intact sites (over 50%), bole accounts for negligible amounts of the above-ground Hg pools (below 0.5%). This is in agreement with reports of very limited translocation of Hg within plants (e.g., Schwesig and Krebs, 2003). Figure 5 shows total ecosystem pools for the four sites with Hg ranging between 44 and  $86 \text{ g ha}^{-1}$ . Most ecosystem Hg resides in soils (upper 40 cm) which accounts for 94 to 98% of the total ecosystem Hg pools across all sites. The notion that soils represent the dominant ecosystem Hg pools has been reported for a number of sites; two boreal forest stands in Canada showed 93 to 97% of the total ecosystem Hg pools to reside in the soils (Friedli et al., 2007), and Engle et al. (2006) reported over 90% of ecosystem Hg in soil in similar Sierra Nevada forest sites and in a desert sagebrush steppe site.



**Fig. 5.** Total ecosystem Hg pools (including soils from 0 to 40 cm depth, left bars; left axis) and corresponding C pools (right bars, right axis) in four Sierra Nevada sites. Error bars represent standard error of pools calculated for individual 4 (3 for Little Valley intact site) replicate plots per site.

There were significant differences in soil Hg pools between the sites located in Little Valley and those near Truckee, and since soils represent the vast pools at each site these differences also show at the total ecosystem level. Soil Hg pools of the Little Valley sites ( $40.6$  and  $41.4 \text{ g ha}^{-1}$ ) are only about 60% of those measured near Truckee ( $67.9$  and  $69.5 \text{ g ha}^{-1}$ ). This is surprising given that all sites are located in climatically similar conditions, in similarly remote locations, at similar altitudes, and all sites experience similar precipitation amounts ( $550 \text{ mm}$  and  $568 \text{ mm}$ ) and regimes driven by wintertime snow. We have no evidence that any of these remote sites were subject to specific point source contributions of Hg. Figure 6 shows that the close relationship between concentrations of Hg to C and N in soils (Fig. 3) also manifests itself by a close association between the pool sizes of C, N, and Hg in soils and hence at the ecosystem level. Soil Hg storage in the top 40 cm calculated individually for the 15 plots of the four sites is strongly related to the respective C and N pools of the plots. The correlation coefficients of linear regressions are strong between soil Hg and C pools ( $r^2=0.84$ ) and – similar to concentrations – relationships are even more pronounced between soil Hg and N pools ( $r^2=0.97$ ). We hence attribute the higher Hg pools present in the Truckee sites predominantly to a higher abundance of C and soil N, although effects of soil texture (as discussed above) cannot be ruled out either. One possible reason for the relationships between soil C and N to soil Hg is that high C and N pools may directly lead to a high sorption capacity



**Fig. 6.** Scatter plot and linear regressions between (a) soil Hg and C pools and (b) soil Hg and N pools. Circles represent plots of the sites near Truckee, CA, and triangles represent plots of sites in Little Valley, NV. Filled symbols are intact sites while open symbols represent post-fire sites (no significant differences).

in soils, which hence leads to a more complete retention of atmospherically deposited Hg in soils. There is, however, a second interesting possibility, that is that high C and N pools reflect a high ecosystem productivity which leads to corresponding high input of atmospheric Hg via plant litter-fall and plant senescence. Litter-fall atmospheric Hg deposition has shown to be important and generally exceeds deposition loads via direct wet deposition, with several forest studies showing an average deposition of  $\sim 16 \mu\text{g Hg yr}^{-1}$  from leaf litterfall alone (not even including any tree senescence) versus  $\sim 8 \mu\text{g Hg yr}^{-1}$  via wet deposition (Bishop and Lee, 1997; Grigal et al., 2000; Iverfeldt, 1991; Munthe et al., 1995; Rea et al., 1996; St. Louis et al., 2001; Lindberg, 1996). Hence, sites with higher productivity exhibit higher atmospheric deposition rates, an effect that may manifest itself by high soil C, high soil N, and high soil Hg concentrations. We believe that both effects (i.e. increased sorption and increased deposition) may contribute to observed close relationships between soil C, N, and Hg levels, indicating that ecosystem Hg pools are determined to a large degree

by ecosystem processes with high organic matter, C, and N pools leading to corresponding large pools of Hg.

A further possibility of higher Hg pools in the Truckee sites, however, could also be due different background levels of Hg in the underlying geologic substrate. To explore this possibility, we estimated geologic background Hg levels by extrapolation of the strong regression of N to Hg to the y-axis intercept in Fig. 3b at each site. Since most soil N is composed of organic N (>95%, Johnson, 1992), the y-axis intercept should theoretically represent the background soil Hg levels in the absence of organic N. We find higher y-intercepts in the Truckee sites as compared to the Little Valley sites (9.7 versus  $4.9 \mu\text{g kg}^{-1}$ ) indicating that the Truckee sites may in fact have higher geogenic background levels. However, multiplication of the geogenic background Hg concentrations with the respective soil masses of the sites (0–40 cm; Truckee:  $2392 \pm 132 \text{ tons ha}^{-1}$ , Little Valley:  $4548 \pm 211 \text{ tons/ha}$ ) yield nearly identical geogenic background Hg pools at each site ( $23 \text{ g Hg ha}^{-1}$  at Truckee, and  $22 \text{ g Hg ha}^{-1}$  at Little Valley). This suggests that different background Hg levels (about a difference of  $1 \text{ g Hg ha}^{-1}$ ) between the sites are not likely to explain an observed large difference in soil Hg pools of 25 to  $30 \text{ g ha}^{-1}$  between the two locations. However, given the different densities and soil textures between the sites it is likely that soil physical properties also may play a role in observed differences between the Little Valley and the Truckee sites.

### 3.5 Effects of fires on C and Hg pools

No significant changes in soil Hg pools between burned and unburned sites were observed at both sites (Fig. 5) which indicates that fires and subsequent changes in vegetation and possibly ecosystem productivity have not – or possibly not yet – affected the underlying soil Hg pools. This is in full agreement with the literature which reports negligible losses of soil Hg during wildfires (Engle et al., 2006). However, above-ground pools at both sites were affected by fires and showed only about half of the Hg pools of the adjacent intact sites (45% and 52% lower for the Lake Tahoe and the Truckee site, respectively; Fig. 4). In the Truckee site, the reason for losses of Hg pools are a reduction in litter pools as the prescribed burning conducted in 2002 removed 67% of the forest floor mass (Murphy et al., 2006) while leaving this pool unaffected in the intact control site. The fire situation in the Little Valley, however, was considerably different: a stand-replacing wildfire in 1981 and salvage logging in the following year removed all trees on one of the two sites while the adjacent intact site had only received some forest floor burning with no or very little mature tree mortality. The difference in Hg pools between the intact and the post-fire site in Little Valley was, therefore, not driven by losses of litter Hg (since this layer was burned at both sites), but by a reduction in above-ground tree biomass at the wildfire site. Because

the post-fire site is dominated by a successional shrub community, the effects are still noticeable in lower Hg pools at this site 20 years after the fire. These results are in agreement with a previous study in the Sierra Nevada Mountains (Engle et al., 2006) which reported that dominant losses of Hg from a prescribed fire originated from the combustion of litter while a wildfire site showed larger Hg losses through removal of both litter and aboveground vegetation. Our study is also in support of many fire studies which indicate substantial losses of Hg to the atmosphere by wildfires mainly due to combustion of above-ground biomass and litter (Obrist et al., 2008b; Sigler et al., 2003; Friedli et al., 2001, 2003a, b; Artaxo et al., 2000; Brunke et al., 2001; Ebinghaus et al., 2007).

#### 4 Summary

Results from our study show that most Hg in these forest ecosystems is retained in the soil and that soil pools are strongly influenced by the presence of soil C and N. These associations are likely the result of a high sorption capacity provided by organic C and N groups which efficiently retain atmospheric Hg deposition, and by high C and N pools reflecting a high ecosystem productivity which also leads to elevated atmospheric deposition inputs via leaf litterfall and plant senescence. Soil N proved to be a particularly reliable predictor for soil Hg levels explaining over 90% of the variance in both concentrations and pool sizes across all measured sites and soil horizons. Various decomposed above-ground litter horizons showed a similar correlation between Hg and N as in soils, but litter Hg was inversely related to litter C concentrations. This could reflect a preferential retention of Hg over C during decomposition, similar to the well-known N/C ratio increases in decomposing litter. A further possibility includes continued and long-term atmospheric Hg deposition to older, well-decomposed litter layers as compared to new, undecomposed litter. Finally, our results show that prescribed burning and wildfires remove significant amounts of above-ground Hg pools, an effect which can persist at sites even 20 years after the occurrence of fires.

*Acknowledgements.* We thank Carsen Berger, Johnny Dagget, Rebecca Higgins, Gabriel Marty, and Sharat Vadwalas for sample collection, processing, and laboratory analyses. The authors gratefully acknowledge Roger Walker and Wally Miller for the use of soil mass and biomass data for scaling up Hg stocks to the ecosystem level. This study was supported by the US Environmental Protection Agency through a Science To Achieve Results (STAR) grant (# RD833378010) to study the consequences of global change for air quality.

Edited by: T. Laurila

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